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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of : BENAZZI et al

Serial No. : 09/909,805

Group Art Unit : 1764

Filed: July 23, 2001

Examiner: Walter D. Griffin

For : PROCESS FOR IMPROVING THE POUR POINT OF FEEDS  
CONTAINING PARAFFINS USING A CATALYST BASED ON A BRIDGED  
DIOCTAHEDRAL 2:1 PHYLLOSILICATE

DECLARATION UNDER 37 C.F.R. § 1.132

Honorable Commissioner  
of Patents and Trademarks  
Washington, D.C. 20231

Sir :

I, Germain Martino, duly warned, declare and say as follows:

THAT, I am a French citizen; that I graduated from "Faculté des Sciences de l'Université de Strasbourg" (France) in 1961; that I obtained an Engineer Diploma from "Ecole Nationale Supérieure de Pétrole et des Moteurs" Rueil-Malmaison (France) in 1963; that I was received as a Doctor by "Université de Louvain" (Belgium) in 1965; and that I now reside in 78300 Poissy (France), 80 avenue Fernand-Lefebvre;

THAT, I was hired by "Institut Français du Pétrole" Rueil-Malmaison (France) in their Research Department to research on catalytic agents and catalytic reactions in May 1967; that, from January 1985 to September 1989, I was Manager of the Kinetics and Catalysis Research Division; that, from September 1989 to December 1997, I was Assistant Manager of the whole Refining and Petrochemical Technology Business Unit; and that since then I have been Manager of said Refining and Petrochemical Technology Business Unit.

THAT, I am familiar with phyllosilicates catalysts and hydroconversion processes.

I declare further:

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**Example 6: Preparation of bridged phyll silicate PDP3 (not in accordance with the invention)**

The phyllosilicate PD1 prepared in example 1 underwent a bridging step using the following operating procedure:

8g of the prepared PD1 phyllosilicate was directly dissolved in 148 ml of a solution of Keggin ions  $[\text{Al}_{13}\text{O}_4(\text{OH})_{24}(\text{H}_2\text{O})_{12}]^{7+}$  having a concentration around  $9 \times 10^{-3}$  mol/l, with a pH of 4. This solution was prepared using the operating procedure published by K. Urabe et al., "Advanced Materials", n° 11 (1991).

The mass of phyllosilicate to be bridged per total solution volume was thus 54 g/l. The ratio between the quantity of polycations engaged multiplied by the charge on the polycation and the quantity of sodium present in the phyllosilicate was 1.

After an exchange period of 7 minutes, with stirring at room temperature, and a filtration step of 1 minute, the product was washed with distilled water for 2 minutes, then dried overnight at 90°C (about 15 hours). The mass of bridged phyllosilicate after ion exchange and drying at 60°C was 0.85 g. The reticular spacing  $d_{001}$  was equal to 1.92 nm. After 5 hours calcining at 550°C under air, the bridged phyllosilicate had a reticular spacing of 1.83 nm and a specific surface area measured by the BET method around 230 m<sup>2</sup>/g. This obtained bridged phyllosilicate was designated PDP3.

**Example 7: Preparation of catalyst C3 (not in accordance with the invention)**

The bridged phyllosilicate PDP3 as described in example 6 was ground with SB3 type alumina provided by Condéa. The ground paste was extruded through a 1.4 mm diameter die. The bridged phyllosilicate content in the catalyst support (phyllosilicate + alumina) was 50 wt %.

The extrudates were dry impregnated with a solution of tetraamine chloride salt  $\text{Pt}(\text{NH}_3)_4\text{Cl}_2$  and finally calcined under air at 550°C. The platinum loading on the final catalyst was 0.6 wt%. The obtained catalyst was designated C3.

**Example 8: Catalytic evaluation of catalyst C3 in comparison to catalysts C1 and C2**

The C3 catalyst was evaluated in hydroconversion of a hydrocracking residue aimed at improving the pour point of the feed. The charge was the same as the one used in Example 5 and is described below:

Sulfur loading (ppm wt)	13
Nitrogen loading (ppm wt)	2
Pour point (°C)	+ 37
Distillation:	
- initial boiling point (°C)	277
- 10% point (°C)	343
- 50% point (°C)	410
- 90% point (°C)	463
- end boiling point (°C)	538

The catalyst was first reduced *in situ* under hydrogen at 450°C before the catalytic test. The reduction was performed using a first temperature step at 150°C for one hour, then increasing the temperature until 450°C with a rate of 1°C/min then staying at 450°C for 2 hours. During this reduction the hydrogen flow was 1000 liters per liter of catalyst.

The reaction was performed under the following conditions: total pressure of 12 MPa, hourly space velocity of 1 h<sup>-1</sup>, hydrogen flow rate of 1000 liters per liter of feed.

The characteristics of the lube oil obtained with Catalyst C3 are indicated in the following table, in which the characteristics of the lube oils obtained with Catalysts C1 and C2 are also plotted, for comparison purpose:

	Catalyst C1	Catalyst C2	Catalyst C3
Viscosity index VI	118	121	115
Pour point	-13°C	-15°C	-11°C
Lube oil yield (wt.%)	68	72	60

This example shows that the use of a catalyst containing a bridged phyllosilicate with a reticular spacing lower than 2 nm leads to performances which are not as good as the ones obtained with a catalyst containing a bridged phyllosilicate with a reticular spacing higher than 2 nm.

The undersigned declares further that all statements are made herein of his own knowledge are true and that all statements made on information and belief are believed to be true ; and further that these statements are made with the knowledge that willful false statements and the like so made were punishable by fine or imprisonment, or both under Section 1001 Title 18 of United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Rueil, April 16, 2003

Germain MARTINO